

DETERMINATION OF CA-41, I-129 AND OS-187 IN THE ROCHESTER TANDEM ACCELERATOR AND SOME APPLICATIONS OF THESE ISOTOPES; U.Fehn, D.Elmore, H.E.Gove, P.Kubik, R.Teng, L.Tubbs, G.R.Holdren Nuclear Structure Research Lab., University of Rochester, Rochester, NY 14627

The TAMS program at the University of Rochester was started about seven years ago at the MP Van de Graaff accelerator (nominal voltage 12 MV). At present we measure several hundred samples per year using roughly a third of the accelerator's up-time. Most of the work is concerned with the determination of long-lived, cosmogenic radioisotopes such as Be-10, C-14 and Cl-36. Examples of investigations recently completed or still in progress are the determination of Be-10 in lake sediments in New York [1], the measurement of C-14 in prehistoric vegetables in North America [2] and the investigation of Cl-36 in meteorites [3]. Two more isotopes were added to this list recently, Ca-41 and I-129, the measurement of which will be the focus of this paper. In addition, we will report on our plans to use the $^{187}\text{Os}/^{186}\text{Os}$ ratio for the differentiation of extraterrestrial material in a meteor crater in Germany.

Calcium-41 has a half-life of 100,000 yrs and is produced in the top few meters of the earth's crust by the interaction of secondary cosmic ray neutrons with Ca-40. After it is released because of weathering it enters the hydrologic cycle and the biosphere. It is thus of potential use for the dating of groundwater as well as of bones in the age range between 50,000 and 1 million years. The expected equilibrium concentrations $^{41}\text{Ca}/\text{Ca}$ are about 10^{-14} [4], which make detection limits of 10^{-15} necessary for these applications.

A major problem for the measurement of Ca-41 with TAMS is the fact that calcium does not readily form negative atomic ions. It does, however, form negative molecular ions such as CaO^- and CaH_3^- . Another problem is the separation of Ca-41 from its isobar K-41, which is quite common in samples as well as in the cesium of the ion source.

One way to eliminate both of these problems would be to use CaH_3^- , because KH_3 apparently does not form negative ions [5]. The probability for formation of CaH_3^- is, however, very low unless calcium metal samples are used. Since the reduction of calcium from small natural samples is quite inefficient and difficult we investigated a different approach, namely the production of negative ions from oxygen bearing Ca molecules. Natural samples (such as bones where Ca occurs as Calcium phosphate) can easily be transformed into CaO or CaCO_3 compounds. We studied the production of CaO^- ions from compounds such as CaO and CaCO_3 , and from oxygen free Ca molecules which were sprayed with oxygen gas. The best results were obtained from CaO^- and from CaCO_3 samples. KO^- ions are, however, present in contrast to KH_3^- ions. The interference of K-41 ions sets a detection limit of approx. 5×10^{-13} for the $^{41}\text{Ca}/\text{Ca}$ ratio under the present conditions.

This detection limit is not sufficient for the measurement of terrestrial samples. Improved chemical procedures during the preparation of the sample and a reduction of the potassium content in the cesium of the ion source will help to lower this detection limit. In addition, we plan to install a new high intensity ion source [6] which should result in a significantly higher ionization rate for the Ca-41 in the samples.

DETERMINATION OF CA-41, I-129 AND OS-187

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Iodine-129 has several characteristics which are outstanding among the cosmogenic radioisotopes, among them its relatively long half-life of 16 m.y. In nature it has two sources: It is produced in the atmosphere by the interaction of cosmic rays with xenon, and in the crust as daughter product of the spontaneous fission of long-lived radioisotopes, mainly U-238. In addition, a significant amount comes from nuclear bomb tests in the atmosphere.

Because of its low natural concentration relatively few data are available concerning production rates and equilibrium values in the various natural reservoirs. The steady-state ratio of $^{129}\text{I}/\text{I}$ in the hydrosphere is estimated to be between 3×10^{-13} and 3×10^{-12} [7,8]. I-129 in this reservoir is derived probably in equal parts from the production in the lithosphere and in the atmosphere [7,9].

These ratios are approx. one order of magnitude higher than the detection limit presently reached with TAMS [10]. A recent measurement of $^{129}\text{I}/\text{I}$ in the Great Artesian Basin in Australia gave values of 5.7×10^{-13} [11] which is in good agreement with the predicted prebomb equilibrium values in the atmosphere. The recent installation of a new injector system will lower the detection limit for I-129 by about one order of magnitude.

A major project we are about to start is the application of I-129 as tracer for hydrothermal convection in sediment-covered oceanic crust. For quite a while now it has been postulated that during the cooling of newly formed oceanic crust seawater penetrates the crust. The reactions occurring between basalt and seawater during this convection change drastically the composition of the circulating fluids and thus may be of great consequence for the element budget of the oceans. Although the most vigorous form of this process occurs right at the active spreading centers, heat flow investigations show that this process is widespread also in older oceanic crust. Because the movement in sediments and older crust is very slow a highly sensitive tracer is needed for the detection of this movement. We plan to use I-129 profiles in conjunction with heat flow measurements in order to determine rate, continuity and extent of this convection in older crust. As a preliminary step of this investigation we are in the process of determining the I-129 content in deep sea sediments from piston cores taken off Cape Hatteras.

Osmium and rhenium are both significantly but not to the same extent depleted in crustal material as compared to extraterrestrial material. This observation combined with the fact that Re-187 is radioactive and decays into Os-187 has led to the suggestion to use the isotopic distribution of osmium as tracer for extraterrestrial material. The Cretaceous-Tertiary boundary is a well publicized example of such an application [12].

We plan to use this approach for the Ries crater in Germany. Although the meteoric origin of this crater has been widely accepted by now it has not been possible so far to identify material of definite extraterrestrial origin. One major obstacle is the dilution of the meteoric material which is significantly higher in the Ries crater than the dilution estimated for the C-T boundary. Calculations of the expected differences in the isotopic composition of Os in the various rock units show, however, that they should be of sufficient magnitude in order to be detectable with TAMS. At present we are in the process of establishing the sensitivity of the accelerator for this isotope system by measuring the Os ratios in artificial samples and in meteorites.

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